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BEHAVIOUR AND BENEFITS OF INTIMATELY MIXED HYBRID COMPOSITES.(U)
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PROPELLANTS, EXPLOSIVES AND ROCKET MOTOR ESTABLISHMENT

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BEHAVIOUR AND BENEFITS OF INTIMATELY MIXED HYBRID COMPOSITES

by

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SUMMARY

Starting from the constant strain theory of hybrid tensile strength, which only describes a lower bound for most hybrid composites, this report develops arguments to show how, in finely-mixed unidirectional hybrids the reinforcing strength of a set of fibres will assume higher values depending on their surroundings. In the case of hybrids of high modulus (HMS) carbon fibre, three characteristic levels of strength are predicted and indeed observed in the experiments reported here. These levels are, the mean fibre strength, the bundle strength referred to short gauge length, and the brittle strength which is also observed in all-HMS composites. Statistical co-ordination solutions are developed which predict the compositions of the average-bundle and the bundle-brittle transitions and also the hybrid tensile strength.

Recommendations are made for several practical systems. Those that have so far been investigated show the predicted trends, of which the most interesting are first, the use of HMS fibres with high tensile (HTS or Toray) carbon fibres to increase the stiffness and damage threshold of complex structures without serious loss of strength and second, the introduction of a glass fibre diluent which increases both work of fracture and strain to break, whilst lowering cost.

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1 INTRODUCTION

Many different types of reinforcing fibre are now in commercial production, and they provide options in strength, stiffness, surface adhesion, density and cost, but no single fibre possesses all the good qualities. It is therefore natural to try mixing them together, especially if one makes the bland assumption that the resulting properties of a mixture will be a linear average of those of the ingredients, the notorious rule-of-mixtures. There has in the past been a good deal of unscientific speculation about this approach, the optimists feeling that the best of both worlds can somehow be attained by "synergism", the pessimists believing that the strength of the whole will be reduced to that of the weakest phase present, a characteristic of poorly-mixed moulding compounds.

Practical evidence has since shown that these extreme arguments do not apply, and recent research has concentrated on distinguishing the limited areas where mixtures of fibres may be used to advantage. The existence at PERME Waltham Abbey of an alignment process which enables fibres to be mixed intimately, rather than in tows or layers, has enabled work to be carried out on ideally mixed systems. Disproportionate changes in tensile strength and work of fracture were found, without compensating losses of interlaminar shear strength. This interim report deals with the results of a first theoretical study, the explanation of the observed facts, and grounds for further work.

1.1 Interpretation of Previous Work

Much of the study of hybrid systems has been carried out using composites of carbon fibre and glass fibre. These two fibres are wholly elastic and possess similar strengths but markedly different strains to break, due to their differing elastic moduli. They also differ in cost by two orders of magnitude. For a woven glass-carbon system, Phillips[1] has shown that the strength of the hybrid is not that of its weakest component. Assuming that his woven bundles of glass and carbon were subjected to equal strains, the hybrid composites failed when the normal breaking strain of the carbon bundle was reached. Bunsell and Harris[2] confirmed that this was so for high modulus carbon and glass, with the carbon fracturing below 0.5% elastic strain as usual. They noted that differential shrinkage should bias the carbon fibre slightly into compression after hot moulding such a laminate. They also showed that the presence of glass bundles deflected or stopped the crack emanating from a failed bundle of carbon fibres. However, the pitch of their glass bundles was perhaps too coarse to

prevent the premature brittle failure in each carbon fibre bundle, which Wadsworth[3] has shown to be characteristic of surface-treated high modulus carbon. Notching experiments[4] have shown also that defects less than 0.25 mm in size will make brittle HMS carbon-epoxy even weaker, which suggests that crack propagation must be controlled on a fine scale if any favourable change is to be brought about.

By mixing continuous HMS carbon fibre and glass fibre more finely, Aveston and Sillwood[5] were able to show that a composite could be made in which the HMS component attained a much higher strain to break, around 1%. However, the carbon was a very small proportion (3.5%) of the whole. They suggested that this unusual behaviour occurred because the carbon was heavily constrained by its surroundings, which prevented release of sufficient energy to break individual fibres. This argument was contrary to the observations of Wadsworth, who was able to measure the breakage of individual carbon fibres, when they were stretched in epoxy resin as a thin constrained layer on a massive aluminium plate. Meanwhile, research at MBB[6] using fine mixing of short fibres, but inaccurate alignment of them, had shown that the flexural strength of some glass composites containing a much larger amount of HMS fibre was also higher than that expected from a theory of constant strain to break. The appearance of the fracture faces showed that the normally brittle fracture of HMS composites had also been inhibited.

During their recent study of the strengths of HMS and HTS carbon fibres, Hitchon and Phillips[7] have noted that the strength utilisation of HMS fibres in orthodox binary composites is poor, in contrast to that for HTS fibres, which is good. We are fortunate to have their results for 5 mm and 0.5 mm lengths to reproduce here as Table 1. Although the effect of length is not large in this case, the observations for 0.5 mm lengths are the most relevant, since stress-transfer through a treated carbon fibre is accomplished in about 0.5 mm. This is also why in discontinuous fibre composites these same fibres, chopped to 2 to 3 mm lengths, will exhibit similar reinforcing strengths to their nominally continuous counterparts. The mean strengths of HMS and HTS fibres are similar, but their different stiffnesses lead to mean breaking strains of 1.08 and 1.7% respectively. A conservative estimate of their potential reinforcing strength is obtained by assuming that once a fibre has fractured it will no longer support any load over about a distance of 0.5 mm.

Using their data (Table 1) for variation in fibre strength, then according to Colemen[8] (Fig 1) this will lead to a maximum load-carrying capability (the bundle strength) at strains of 0.78% for HMS and 1.23% for HTS respectively. The latter value is close to the accepted reinforcing strength of HTS fibre, the former however is nearly twice that actually observed for the HMS fibre used here.

Thus Aveston's and Sillwood's[5] observation of HMS apparently breaking at about 1% strain can instead be interpreted as first, the absence of a load maximum since the very high proportion of glass fibres continue to accept load and to frustrate brittle fracture; second, the proportion of HMS was small enough that significant departure from linear elastic behaviour would only be seen when nearly half the HMS fibres were broken to lengths incapable of stress-transfer, ie at the mean breaking strain for HMS of 1.08%.

The problem of very brittle failures recurs with some regularity in the use of carbon fibres. The need to ensure that interlaminar shear strength is sufficient for design purposes made it essential for surface treatments to be introduced to improve adhesion, but they are not easy to control accurately, and when carried to excess, they do cause premature fracture. At the time of writing, some very high strength carbon fibres only exist in such a form, with highly active surfaces, so that an understanding of hybrid behaviour may be important here.

1.2 General Approach

The problem of predicting accurately the mode of fracture in a composite and the transition from wholly consecutive fibre fracture to the more desirable, random fibre fracture has taxed many authors,[9] and precise estimates are very difficult to make. However, given the existence of fibre composites which do fail prematurely various practical solutions have been proposed to improve them, one of which is to blend these fibres with those that make a tougher composite. In this case it is suggested therefore that two useful mechanisms exist:

a Premature brittle fracture of successive fibres can be postponed provided it is done on the correct scale. This is largely a topological condition.

b Then if the load they carry can be taken up, a large proportion of the fibres of lower breaking strain can be broken before there is an observable departure from elasticity, and the net strength of the hybrid

will be that due to combining two sets of fibres, ie the bundle strength of the two groups taken together.

Some expected practical advantages in discontinuous hybrids would be first to compensate for poor packing by introducing some HMS fibre into HTS materials, and then to make composites which are more tolerant of defects and damage generally, and in which the reinforcing strength of certain fibres is enhanced.

The difference in fracture behaviour between HMS and HTS discontinuous fibres aligned in epoxy is clearly seen from their fine-scale work of fracture, which we have measured by the method of Tattersall and Tappin (see Appendix C). In spite of their similarity in fibre strength, there is at least a fourfold difference in work of fracture of the composites. This is a relevant measurement since in engineering use, composites must tolerate the presence of fine defects which cannot be detected by inspection. The machining of simple tensile test pieces will in any case introduce defective regions much larger than the fibres themselves. Any theoretical model of hybrid behaviour must therefore consider the propagation of a defece (or lack of it) as a mechanism controlling the observed strength. By extrapolating from the effect of small notches[4] a defect size of only 36 μm would be sufficient to cause fracture at 1% strain in an HMS composite, in the absence of other causes. The corresponding figure at the bundle strength of HMS (0.78%) would be 60 μm (assuming that elastic strain to break varies inversely with square root of defect size).

If the HMS fibres are prevented from becoming adjacent to each other then there is no reason why premature failure should occur. Thus in a highly ordered system at a ratio of one HMS fibre to two HTS fibres, assuming hexagonal close packing, each HMS is on average fully co-ordinated by HTS fibres. If the ratios are reversed then the HMS network forms a virtually continuous structure (Fig 2). Unfortunately, it is not yet possible to produce such a systematic, finely-mixed structure, and so an estimate must be made for randomly mixed, but highly aligned fibres.

2 DEVELOPMENT OF THEORETICAL MODELS

2.1 Lower Bound for Hybrids Containing Fibres or Layers of Low Variability

For simplicity, we allow the total proportion of fibre in a composite to remain fixed, at say 50%, but vary the relative volumes of the two fibres to be mixed. The properties of the two end fibre-resin systems are known. If the hybrid consists of a mixture of two fibres, A and B, the stiffness of B typically being greater than A, but its breaking strain less than that of A, then a working diagram can be constructed to predict the strength of the system, Fig 3. Lines of equal elastic strain can be projected from the origin O, chosen so that OA/OB = modulus A/modulus B. The elastic moduli for hybrid compositions then lie along a strain line such as EE'. Thus if an all-B composite fails at stress P, then PR is the line of constant strain along which the B fibres would be expected to fail. If S represents the strength of the all-A composite, SB represents the contribution of A to the hybrid strength. It follows from the simple strain argument that between P and T, breaking of the B fibres will precipitate complete failure. For compositions to the left of T the A fibres should be capable of taking the extra load when the B fibres break. The A fibres can then be loaded further until the line SB is reached. Consequently this strain argument gives the line PTS as defining the strength of the hybrid system.

If failure of the B fibres causes a serious stress concentration, ie a defect which is above critical size, then total failure may persist along TR. A defect which is just sub-critical at stresses TR may still become critical and lead to failure below TS values.

By setting down the expressions representing PR and BS, we can show that the strength minimum at T is related to composition by

$$a_T[1 + R_E\,(R_S - 1)] = 1, \text{ assuming elastic behaviour}$$

where a is the relative proportion of A, $R_E = \frac{E_A}{E_B}$, the ratio of the moduli, and $R_S = \frac{\varepsilon_A}{\varepsilon_B}$ the ratio of the breaking strains of compositions A and B respectively.

Similarly the depression of strength at T relative to the rule of mixtures is

$$\frac{S_M}{S_C} = 1 + \frac{R_S - 1}{R_S}$$ and therefore is never less than half rule of mixtures.

These results are important when strength is to be traded off for other factors, eg energy absorption on a rising load curve, which occurs along TS as the B fibres break, but not along TP. Here complete fracture occurs as the B fibres fail, although the energy needed to cause the fracture may well be higher than at P. (The energy absorbed on a rising load curve cannot, of course, exceed twice the total elastic energy of the A component alone.)

The extent to which the depression of strength along STP can be reduced by intimately mixing real, variable fibres is discussed below and in Appendix A.

2.2 Mixed and Variable Fibres (the Average-Bundle Transition)

The above model applies where each set of fibres is very consistent. If there is statistical variation in the strength of the fibres of type B for example, then their strength contribution realised between T and S in Fig 3 will apparently be enhanced, ie because no load maximum is then reached in breaking type B fibres, progressive failure will occur, and their strength at least to the left of W will be characterised by the average value, eg from V rather than P. In principle, an apparent strength increase of B by some 40% could occur. Fig 4(a) illustrates this problem of calculating strength for real, variable fibres in a system where the strains to break of A and B overlap. This can be treated arithmetically by weighting the distributions A and B for their relative stiffness and proportions, then adding them to identify the strain corresponding to maximum load, as successive fibres fail. Alternatively, the combined reinforcing strength may be estimated using Coleman's relationship for coefficient of variation (C) and bundle strength as a proportion (ρ) of the overall mean strength (Fig 1).

Covariance statistics appear to have the right properties for solving this problem algebraically, ie assume overall mean strain to break is

$$X = p\,x_A + q\,x_B \qquad (1)$$

where x_A and x_B are the separate average fibre strains, p and q the weighting factors, then combined variance (where $C^2 x^2 = V$) is

$$V = p^2 V_A + q^2 V_B + V_C \qquad (2)$$

and this expresses the variance due to each distribution (V_A, V_B) and to the

spacing between them (V_C). Clearly this expression has limited application. The total distribution is not usually normal and the treatment breaks down when the spacing between individual distributions is large, so that each is broken separately, Fig 4(b). However, under these conditions, the effective strength fortunately is self-evident. Application of Equations 1 and 2 is discussed in Appendix A.

2.3 Suppression of Premature Matrix Failure

Aveston, Cooper and Kelly[10] have derived an expression for the constraint imposed on the brittle cracking of a matrix by reinforcing fibres, and Aveston and Sillwood suggest that this might be extended to a stiff matrix containing other brittle elements, such as HMS fibres. Such a theory may be used to predict whether or not this brittle matrix will crack before the main reinforcing fibres fail. However, it is implicit in their arguments that the A type fibres of Fig 3 eventually bear all the load. Thus their theory is useful for predicting the shape of stress-strain curves in the region below TS, but can do little to predict the tensile strength of other compositions, particularly when some of the main reinforcing fibres have already been broken. However, extra energy absorption on a rising load curve is a useful feature which is worthwhile developing and their predictions for practical systems are discussed in Appendix B.

3 CO-ORDINATION HYPOTHESIS FOR MIXED FIBRES OF SIMILAR DIAMETER

The essential problem in dealing with premature brittle fracture of fibres within a unidirectional hybrid is to decide which fibres are subjected to it, and which are not. Although fractures spread quickly through an all-HMS system, will this be so if each HMS or B type fibre is surrounded by "tough" composite composed of A type fibre? It is asserted that the B fibre will then in general be protected. Therefore an attempt has first been made to explore the environment surrounding B fibres assuming a co-ordination number of five, which is most characteristic of normal, imperfect fibre packing.

For a CN of five, there are six possible ways of surrounding a fibre, ie 5As, 4A + 1B, 3A + 2B, 2A + 3B, A + 4B, and 5B. If A and B are in the relative proportion a, b = 1 - a, then the probability of selecting 5As is a^5, similarly that of selecting 5Bs is b^5, and the probabilities for the other conditions are

available* from the expansion of the binomial $(a + b)^5$. The probability coefficients for these symmetrical expressions are given in Table 2, and the characteristics they represent are:

5As	an isolated B
4A + 1B	the end member of a B Chain
3A + 2B	part of a single chain or Bs
2A + 3B	part of a double chain, a junction of Bs
A + 4B 5B	part of a large block of Bs

Thus for values of a = 0.9, b = 0.1, the B fibres are largely isolated, and the A fibres a virtually continuous block, but in the range 0.4 to 0.6 the mixture consists largely of single and double interwoven chains. Under these conditions one can visualise that consecutive cracking of B fibres could proceed along chains, but that similarly, a single devious chain of A fibres might eventually run right through the thickness of a sample and be used to frustrate crack propagation by providing a weak boundary which can fail in shear. We also note that when b > 0.8, the number of chain end members is very small, suggesting a continuous extensive structure of B.

3.1 Composition Range of the Bundle-Brittle Transition

The object of this section is, with the least assumptions and preferably with the least calculation, to bracket the estimated composition range in which crack propagation through the B phase ceases to be inhibited by the A. Excluding all other factors local variations in composition will of course prevent the existence of a precise transition. However, we first assume that the average proportions of A and B are maintained in the volumes to be considered, and attempt a calculation for thin sections, eg shells, laminations and indeed the strip test specimens used in this work. This is shown in Figure 5 which illustrates propagation from a through-thickness edge notch or pre-existing path of Bs. Propagation of cracking through the B phase is presumed to take place where consecutive B fibres exist, but to be frustrated by the presence of a line or path of A fibres. Thus for continuous cracking, the proportion of A

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*See for example M J Moroney, Facts from Figures, Penguin Books

(a_s) needed to create a single blocking line must be greater than that of A which actually exists when propagation through B is occurring, ie if b' is the critical proportion of B at which brittle cracking can first occur, then $a_s > (1 - b')$. Now the composition for a single line of A must be numerically the same as that for a single line of B, ie $a_s = b_s$, so

$$b_s > (1 - b') \tag{4}$$

but $b_s < b'$, since the existence of occasional single lines of B is insufficient to sustain crack propagation, hence

$$b' > (1 - b') \text{ and therefore } b' > 0.5$$

The total number of fresh lines or paths available during crack propagation from a single line of B can be estimated directly. Each B consecutively cracked has continuous paths through it, ie for every B surrounded by four other Bs, three new paths are introduced, in a rapidly expanding number of routes. Considering a crack passing through an additional population m of mixed fibres, then at the critical composition b', using the notation of Table 2, the total number of paths involved is no greater than

$$1 \times 2^{C'_4 b'm} \times 3^{C'_5 b'm} \times 4^{C'_6 b'm} = Q' \text{ say, or}$$

$$\log Q' = b'm\,(C'_4 \log 2 + C'_5 \log 3 + C'_6 \log 4) \tag{5}$$

(This is an over-estimate of the number of paths, since some must rejoin.)

Now the ease with which such continuity of B occurs, either in single paths or in multiple paths, is governed by the proportion of chain-forming fibres available, and the number of members required in each chain, as well as the size of the original population from which they are chosen. The proportion of chain-making Bs available is $b(1 - C_1 - C_2)$ and the probability of selecting r fibres consecutively, to form a chain is therefore $b^r (1 - C_1 - C_2)^r$. Thus the number of chains length r is

$$Q_r \propto b^r (1 - C_1 - C_2)^r \tag{6}$$

In the case of single paths of A or B through the thickness of a laminate, r has a characteristic value larger than, but related to, the thickness expressed in fibre diameters, ie $r \approx 10^3$ (which also allows the inclusion of at least one very weak fibre from the strength distribution).

Applying the two expressions (5 and 6) to the special case where, during the initial stage of propagation from a path of r fibres, Q' is changing quickly, but r is virtually constant, gives

$$\left[\frac{Q'}{1}\right]^{\frac{1}{r}} \geqslant \frac{b'\,(1 - C'_1 - C'_2)}{b_s\,(1 - C_1 - C_2)} \qquad (7)$$

also provided $b_s > (1 - b')$.

We take Q = 1 for the case of a single path, for although a single path has many related spurs, these will not in general be of sufficient length (r) to qualify. This is the distinction used here between isolated paths and the continuous paths supporting crack propagation. These two inequalities (7 and 4) can be used to generate acceptable values of b_s, b' from different values of the ratio $\frac{m}{r}$. That is

$$\frac{m}{r}\, b' \; (C'_4 \log 2 + C'_5 \log 3 + C'_6 \log 4)$$

$$\geqslant \log\,[b'\;(1 - C'_1 - C'_2)] \; - \; \log\,[b_s\;(1 - C_1 - C_2)]$$

and $b_s > (1 - b')$, interpolating the coefficients in Table 2 as necessary.

Now the difference between b_s and b' becomes small as m diminishes, ie

$$\frac{m}{r} \geqslant 2 \qquad b' = 0.60,\ b_s = 0.40$$

$$\frac{m}{r} \geqslant 1 \qquad b' = 0.56,\ b_s = 0.44$$

$$\frac{m}{r} \geqslant 0.1 \quad b' = 0.503,\ b_s = 0.497$$

$$\frac{m}{r} \geqslant 2.6 \quad b' = 0.7,\ b_s = 0.3$$

The accuracy with which a transition can be predicted by this means therefore depends on the increment value of m assumed. There appears to be no reason

why m should not be of the same order as r.

It is perfectly possible that a deeper investigation of the theoretical model would determine a narrower transition range than this, but such precision is unlikely to occur in real compositions.

TABLE 2

The Weighting Effect of Relative Proportions of A and B on Possible Ways of Surrounding a Single B Fibre with Five other Fibres

Composition		C_1 (5A)	C_2 (4A + B)	C_3 (3A + 2B)	C_4 (2A + 3B)	C_5 (A + 4B)	C_6 (5B)
a	b						
1.0	0	1.0	-	-	-	-	-
0.9	0.1	0.59	0.33	0.07	0.01	-	-
0.8	0.2	0.33	0.41	0.20	0.05	0.006	-
0.7	0.3	0.17	0.36	0.31	0.13	0.03	-
0.6	0.4	0.08	0.26	0.35	0.23	0.07	0.01
0.5	0.5	0.03	0.156	0.31	0.31	0.156	0.03
0.4	0.6	0.01	0.07	0.23	0.35	0.26	0.08
0.3	0.7	-	0.03	0.13	0.31	0.36	0.17
0.2	0.8	-	0.006	0.05	0.20	0.41	0.33
0.1	0.9	-	-	0.01	0.07	0.33	0.59
0	1.0	-	-	-	-	-	1.0

This probability argument of stemming crack propagation is therefore summarised as follows. Although it is theoretically possible to suppress at source premature brittle failure in a fibre-matrix combination, by reducing interfacial adhesion, this is sometimes difficult to arrange. If the problem is to be overcome by blending in a "tough" fibre of similar diameter, then for the test specimens commonly in use, it appears that cracking can indeed be suppressed with sufficient frequency, the main topological transition occurring fairly sharply at about equal proportions of brittle and tough fibres, even where the function of the "tough" fibre is purely to provide a boundary of low shear strength, eg polymeric fibres would do, if they were weakly adhering ones.

The strength of compositions nominally on the transition value will be difficult to predict and more variable than usual. On the tough side of the transition, it is proposed that the bundle strength of the brittle fibres would be a guide to their behaviour, following Equations 1 and 2. The concept is illustrated in Fig 6. On the "brittle" side of the topological transition, the local and average load-carrying potential of the tough fibres needs to be considered. If they can support the reinforced matrix, then the Aveston-Sillwood model must be re-examined. If not, the whole will fail at a similar strain to that of the original brittle fibre-matrix combination, with only small corrections for changes of stiffness and work of fracture.

It is amusing to note that a similar transition would be predicted by the enigmatic statement that "it takes two A fibres to protect one B".

3.2 Effect of Fibre Bundles, Large Fibres, etc

The model for crack suppression can in principle be extended to fibres of differing diameters. If large "brittle" fibres exist, these can each be coated with only a small proportion of finer, but tougher ones, equivalent in the limit to modifying adhesion at source. Where the roles are reversed, the result does not seem attractive. A practical realisation of large brittle, small tough would be to deposit in an alignment plant a heavy feed of small brittle fibres at a coarse pitch, to be followed by a light feed of the toughening fibres, which should be effective, even against a major proportion of brittle fibre.

4 EXPERIMENTAL

4.1 Programme

A programme of measurements with a range of hybrid compositions was drawn up, taking account of both the need to validate a theory and the likely materials requirements in MOD, as follows:

1 For thin skins and complex shells, high stiffness materials of adequate ILS, but increased tolerance of damage, including that introduced by stretching felts longitudinally to final shape.

2 HTS materials as in 1.

3 Indigenous replacements for Kevlar, when it is used primarily for damage tolerance rather than for specific tensile strength alone - where it is

now being matched by improved carbon. A significant advantage over Kevlar in compression could also be expected.

4 Upgrading 50% HTS discontinuous to approach 60% HTS continuous in stiffness.

5 Ultimately, cost-effective use of the cheaper fibres, eg pitch fibres, glass, asbestos, and possibly cellulose.

Mixes	Proportions of Composite Volume					Function
HMS-HTS	50-0		25-25	12.5 - 37.5		4
HMS-Toray or HTU		40-10	25-25		0-50	4, 1
HMS-Glass		40-10	25-25	12.5 - 37.5	0-50	3
HTS-Glass	50-0	40-10	25-25			2, 3

So far only the HMS-HTS system has been assessed in any depth, although there is some further indication for HMS-glass from a collaborative programme with MBB Munich, and with NPL. Virtually all the data is encouraging and favours careful completion of the programme as soon as possible. The test methods used are described in Appendix C.

4.2 HMS-HTS Carbon Mixtures (in Epoxy CIBA 914)

In this case a fibre with very brittle behaviour is introduced in a less-brittle system. A master diagram has been prepared for this system and the test results placed on it (Fig 7, Table 3). The flexural stiffness measurements proved more consistent than those in tension and showed the expected linear relationship with composition, which confirmed that the total volume loading of fibre in each laminate had been held close to 50%. Ten specimens were tested for each tensile point recorded. Only in one sample did the measured load-extension curve show non-linearity after the take-up stage was complete. This sample (1 HMS, 3 HTS) was at the top end of the strength distribution and was presumed to be showing the beginnings of progressive break-up of the HMS fibre, about its mean breaking strain.

Corresponding to the linear elastic behaviour, the tensile strengths of the mixtures tested were all well above those expected if the HMS were to break prematurely at its usual strain with the HTS eventually accepting all the load.

The results for the two forms of HTS (75%)/HMS (25%) in fact were slightly above the strain level for the HMS bundle strength, as would be expected from Appendix A. At equal parts of HMS and HTS the breaking strain was 12% below the bundle level. However, HTS-epoxy systems cannot be regarded as completely tough and the stiffness data suggest that the relative proportions could possibly have been nearer 55/45. Measurements of the work of fracture also show that at these proportions the behaviour is more brittle, in contrast to the values for about 25% HMS in the two forms of HTS and in glass (Fig 8), where little reduction is observed from that of the simple HTS or glass composite, ie the "protected" HMS makes its own significant contribution. The interlaminar shear strengths of all the carbon mixtures were consistently high (Table 4).

Both the unidirectional flexural strengths themselves, and the tensile threshold levels after impact in flexure of the 25% HMS crossplies, were high relative to pure HTS (Table 5). This can be expected from the local break-up of the HMS absorbing energy at the high strains experienced in the surface layers of a flexural or slow impact test. Predictably, this effect does not persist at 50% HMS.

Overall these observations tend to support the topological view adopted and for other compositions of HMS/HTS one would therefore expect the following:

a Observable break-up of HMS at strains in excess of 0.8% in the composition range 0 - 20% HMS (100 - 80% HTS), ie useful yielding, with bundle strength predicted by Equations 1 and 2.

b Elastic strain to break levels about 0.8% persisting from 20% to at least 40% HMS/60% HTS, and to about 50% HMS given lower surface treatments, eg in the HMS/HTU or Toray systems.

c Beyond 55% HMS, brittle behaviour, with the upper bound of tensile strength possibly predictable by assuming a constant defect size (c) and correcting for differences of both work of fracture (γ) and stiffness of the mixtures compared with pure HMS, ie breaking strain proportional to $\left(\frac{\gamma_c}{E_c}\right)^{\frac{1}{2}}$. Observations suggest that the measured work of fracture is not fully effective in this way.

4.3 Glass - HMS

There is no definitive work available yet for this system, but its qualitative behaviour can be anticipated. This system is sketched in Fig 9, assuming that the glass fibre does not adhere to the matris as strongly as HTS or HMS fibre, but is correspondingly longer, to enable it to develop adequate reinforcing strength. Then a first minimum in strength should be reached about 28% HMS. Between 0 and 18% HMS it seems safe to assume that the yielding break-up of HMS will occur, as observed by Aveston, and this is a useful composition since the 18% mixture has virtually twice the stiffness of GRP alone. Theoretically then a maximum strength (close to rule of mixtures) should be observed about 45% HMS, and in this case since the glass is not sufficiently strong to prevent consecutive failure of HMS in single chains, the strain to break must fall below 0.8% by 55% HMS, by which stage the net strength contribution from even the brittle HMS phase is greater than that of the glass. The glass can still contribute markedly to work of fracture, however, so forecasting becomes very speculative. If the increased work of fracture is fully effective, the strength of brittle HMS will rise sharply with small additions of glass (U). If, as seems more likely, the HMS assumes the strength of a single layer sheet up to say 0.6 of HMS, the hybrid strength may well oscillate, with the HMS breaking before the toughening effect of the glass comes into play (L).

The evidence from the work of MBB does not contradict the argument, but their measurements were of flexural elastic behaviour, on composites the stiffnesses of which indicate were not accurately aligned. These were pressure moulded, thereby causing frequent fibre to fibre contact down the length of individual fibres. Expressing their very consistent results as strains rather than stresses (Table 6) shows that the HMS was extremely brittle in this condition, failing at 0.29% strain in bending, but attaining 0.85% with 67% glass, 0.65% with 50% glass, 0.43% with 33% glass. They noted that the same fibres prepared in alternate layers of glass and carbon gave lower figures, although these too exceeded worst case predictions. Zweben[12] discusses the need then to take account of the strength of thin layers rather than individual fibres, and treats this aspect using Weibull rather than normal statistics. Finally we note that the adhesion of glass to epoxy was relatively high in the MBB composites (66 MPa) which suggests that the glass had some surface treatment.

5 CONCLUSIONS AND FURTHER WORK

1 The constant strain-to-break theory of hybrid strength explains the majority of observations on coarsely-structured hybrids.

2 With a hybrid mixture, the set of fibres having the lower strain to break can and does exhibit different levels of strength, depending on their environment. This encompasses finely-mixed hybrids and some unusual observations made in the past on hybrid laminates. HMS fibres for example display at least three distinct levels:

a when they are so well supported that they attain or exceed the average strength of HMS fibre (about 1.1% strain),

b when they contribute significantly to the hybrid strength but brittle failure is suppressed and they therefore attain their bundle strength referred to a short fibre length (about 0.78% strain), and

c where consecutive brittle fracture is not suppressed and their strength contribution is similar to that of an orthodox HMS composite (0.4% strain or less).

3 A statistical co-ordination argument has been developed to identify the composition of the brittle-bundle transition for fibres of equal diameter and this is supported by available data, but the introduction of, for example, weakly-bonded polymeric fibres could clarify the de-coupling mechanism suggested by this argument.

4 A statistical weighting method is derived for calculating the strength of non-brittle hybrids when the distributions of fibre breaking strain overlap. It is difficult to disprove this approach within the accuracy of any one set of tests. However, the limits of validity of the algebra can now be explored by using a computer to carry out numerical solutions with artificial data. Graphical methods are given here for preparing estimates of practical systems.

5 The most important practical conclusion is that existing results are within expectations and that further fruitful systems should be investigated as soon as possible. It has already been demonstrated how to increase stiffness (HMS/HTS) without serious loss of strength or work of fracture, and how to increase the apparent strength and work of fracture of strong and expensive

fibres. These results are based on randomly mixed fibres of similar diameter. If engineering solutions can be devised to produce fine, but more systematic grouping, then even more effective compositions can be developed. Optimum hybrids of three or more types of fibre still remain untapped and a more precise understanding of mixing size-effects might also enable continuous fibres to be used successfully. Fatigue strength should be measured to confirm that the advantages are not ephemeral.

6 ACKNOWLEDGEMENTS

Dr H Edwards and Mr N P Evans prepared all the highly aligned felts used in this research, as part of a joint programme to improve the design of complex composite structures.

Mr J Cook has been very generous with his time in helpful discussions.

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APPENDIX A

TENSILE STRENGTH OF TWO SETS OF FIBRES OF VARIABLE STRENGTH

Where the distributions of breaking strain in a mixture of two sets A and B of fibres overlap, it is proposed they be treated as a single distribution[11] with a new weighted mean

$$X = p\,x_A + q\,x_B \tag{1}$$

where $p + q = 1$

so that the new variance is $V = p^2 V_A + q^2 V_B + V_C$ (2)

$$\text{where } 2V_C = p(X - x_A)^2 + q(X - x_B)^2 \tag{3}$$

there being in effect p readings at x_A, q readings at x_B since p and q are weighting factors which allow for the respective stiffnesses and proportions of A and B, ie $p \propto a\,E_A$, $q \propto (1 - a)E_B$. (The procedure could in principle be extended to any number of sets.)

We then use Coleman's relationship (Fig 1) which relates mean strength ($X\,E_C$) to the reinforcing strength of a bundle, through the variation of strength (C) taking $C^2\,X^2 = V$ where $E_C = aE_A + (1 - a)E_B$.

Now for $p + q = 1$, normalising p gives

$$p = \frac{aE_A}{aE_A + (1 - a)E_B} = a\,\frac{E_A}{E_C} \qquad \text{similarly } q = (1 - a)\,\frac{E_B}{E_C}$$

So from (1) $X\,E_C = aS_A + (1 - a)\,S_B$ where $S_A = x_A E_A$, $S_B = x_B E_B$

This approach is therefore equivalent to taking a simple proportional average for fibre strength, but recognizing that the distribution is in general widely scattered with respect to strain and therefore allowing this scatter to determine the strength of a bundle. By inspection it can be seen that p, q, E_C, and XE_C, and therefore X can be determined graphically for any value of a, given the basic fibre data. This has been done for the system glass-HTS/HTU/Toray in Fig 10 and the calculated strength is compared with simpler expectations in Fig 11, using values $x_A = 0.04$, $x_B = 0.017$ with coefficients of variation 0.15, 0.3 respectively. (For convenience in plotting, Imperial units were used.)

This method predicts values for strength which lie between "rule of mixtures" expectations and the strength minimum enforced by assuming a constant breaking strain. Treating the glass as a diluent of lower cost, it appears that relative proportions of 50 - 60% glass with HTS will provide a composite which still has useful stiffness, good utilisation of fibre strength and potentially some increase in damage tolerance compared with HTS without severe degradation as the glass becomes affected by the environment. However, it is not expected that fine mixing would confer much advantage in this system, except at high glass contents. Elsewhere the theoretical differences are sufficiently small to pass undetected.

BREAKING STRAIN OF A REINFORCED MATRIX

Enhancement of the strain to failure of brittle matrices is observed in the direction in which they are reinforced. The increased breaking strain of brittle rosins, cement, glasses and ceramics has frequently been demonstrated and this is most noticeable when the fibres are fine and provide an extended surface, to which the matrix can adhere, thereby preventing the opening of matrix cracks. The critical energy balance[10] which determines the strain at which a crack bridged by fibres will propagate through the matrix phase predicts the critical strain (X) as

$$X^3 = \frac{12\tau\gamma_M V_F}{E_C E_M r} \times \frac{E_F V_F}{E_M V_M} \qquad (8)$$

where the subscripts C, M, F denote composite, matrix and fibre, E being stiffness, V - volume fraction, r - fibre radius, γ - work of fracture, τ - shear stress for slip of matrix past fibre, which tends to open a matrix crack.

Aveston and Sillwood[5] argue that τ is not commonly known, but the stress-transfer length (L) between fibre and matrix is, and it is related to the stress in a fibre by

$$X E_F = \frac{2\tau L}{r} \qquad (9)$$

so if the surface of a matrix crack is strain-free, then (9) may be substituted in (8) which simplifies to

$$X^2 = \frac{6\gamma_M V_M}{L E_C} \alpha \frac{V_F E_F}{E_M} \qquad (10)$$

where $\alpha = \frac{E_F V_F}{E_M V_M}$

To apply this expression (10) to a hybrid containing a set of fibres which break with the matrix by brittle fracture, they must be regarded as part of the matrix, contributing to its stiffness and to its work of fracture, the other (tough) set of fibres acting as the only true "fibres" in the hybrid.

Now γ, the work of fracture for say HMS-epoxy composites, is in the range 0.15 - 0.5 J/cm^2 depending on the sources of resin and fibre used. The value for L will vary from 0.5 to 1 mm for glass down to 0.1 mm for carbon. Using

the data in (3), it predicts that for the HMS-glass and HMS-HTS systems, this form of matrix cracking is very sensitive to relative composition. Cracking will not occur at 1% strain below 30% of HMS with glass or below 90% HMS with HTS, and indeed it has not so far been observed. Aveston and Sillwood examine the similar case where the resin does not crack, but the brittle fibres do.

However, either approach must assume that cracks are bridged and that the set of "tough" fibres therefore controls the strength of the whole system, all the "brittle" fibres set being vulnerable to consecutive cracking, regardless of their immediate environment. The observed strengths and the topological argument presented in this report, suggest that this calculation cannot be used to predict composite strength, but it could prove helpful in estimating departures from elastic behaviour, if the condition for "matrix" cracking were determined by assigning some of the "brittle" fibres to the tough role.

APPENDIX C

TEST METHODS

The following tests were used to assess the properties of each hybrid type, they were designed to use two standard thicknesses of laminate 1 mm and 2 mm, and to minimise as far as possible the amount of prepreg material required.

1 Flexural Strength and Modulus

The standard testpiece[13] was used with a 2 mm thick laminate and 40:1 span to depth ratio (9 samples/board).

2 Interlaminar Shear Strength

The standard testpiece[13] was used with a 2 mm thick laminate and 5:1 span to depth ratio (8 samples/board).

3 Tensile Strength, Modulus and Strain to Break

A 1 mm thick unwaisted tensile specimen was used so as to test the largest volume of material as simply as possible. The specimen was 16 cm long with 4 cm Al alloy plates bonded at each end to spread the gripping loads. These specimens were tested in wedge grips set up so that the greatest clamping force on the specimen was at the ends of the Al plates, reducing to zero at the gauge length, and to ensure that the specimens sat centrally and level in the grips to give correct alignment. The grips ran in a lubricated aluminium trough to ensure accurate parallel motion of the grips and reduce extraneous forces as much as possible.

Strain to break was measured with an Instron strain gauge extensometer, held in place with a thin layer of soft rubber deposited on the specimen surface from rubber solution. This arrangement provided enough friction to hold the extensometer in place during tests (the anvils of the extensometer could not grip the specimens by themselves, and in any case could not have been allowed to bite into the specimen) but allowed slippage at fracture when a more rigid system could have been smashed. The extensometer proved to be quite robust and was undamaged after many specimens had broken and splintered between its anvils (10 specimens/board).

4 Work of Fracture

The specimen was basically that used by Tattersall and Tappin[14] with the notches cut by a 0.2 mm diamond wire saw, but 2 mm thick by about 2.5 mm wide and 12 mm long - tested in an ILSS rig. The size was chosen to be representative of fine defects and allowed material from the board made up for flexural testing to be used. Tattersall and Tappin reported a size effect with their samples; about 6% increase in work of fracture being shown for doubling the size of the specimen. However, the convenience of using standard thicknesses of material outweighs any apparent changes in work of fracture due to sample size. 10 samples were used for each composition. The values given are for total energy and must be halved to derive the energy per crack face.

5 Residual Tensile Strength after Impact

The specimens for this investigation were 12 cm long by 2 cm wide and 1 mm thick of $0^{o}/90^{o}$ balanced crossplies with 4 cm long Al plates bonded at each end, giving a 4 cm × 2 cm gauge length. The samples were impacted by drop weight at 0.5, 1 and 1.5 joules (2 samples/impact energy level) and subsequently tested in tension to obtain a residual strength figure, following the procedure of Reference 15.

TABLE 1

Strength of 50 mm, 5 mm and 0.5 mm Long Carbon Fibres

OBSERVED (AERE) / ASSUMED FOR THIS WORK

Fibre Type and Length	Mean Strength MPa × 10^{-2}	Number of Tests	Standard Deviation MPa × 10^{-2}	Coefficient of Variation %	Mean Fibre Diameter μm	CV Assumed	Bundle Strength as Fraction of Average (Fig 1)	Average GPa	Average % E	Bundle Strength (% E)
A 50 mm										
Type 1 (15BR)										
1st Series 2nd Series	27.1 24.7	121 24	5.66 4.29	20.9 17.4	7.89 8.78	20	0.68	2.6	0.68	0.46
Type 2 (11R)										
1st Series 2nd Series	30.7 27.5	125 25	4.57 3.68	14.9 13.4	7.93 8.90	15	0.73	3.0	1.24	0.91
B 5 mm										
Type 1 (15BR)										
1st Series 2nd Series	41.2 29.4	10 10	7.41 7.07	18.0 24.0	8.13 8.46	20	0.68	3.5	0.92	0.62
Type 2 (11R)										
1st Series 2nd Series	37.1 36.3	10 10	6.87 5.24	18.5 14.4	8.79 9.52	15	0.73	3.65	1.51	1.10
C 0.5 mm										
Type 1 (15BR)										
1st Series 2nd Series	43.6 37.8	10 10	7.10 5.94	16.3 15.7	8.13 8.46	16	0.72	4.1	1.08	0.78
Type 2 (11BR)										
1st Series 2nd Series	48.3 34.6	10 10	7.45 8.78	15.4 25.3	8.79 9.52	15*	0.73	4.1	1.69	1.23

*First series only

TABLE 3

Strength and Stiffness of Unidirectional HTS-HMS Carbon-Epoxy Hybrids in GPa, 50% Fibre by Volume, Coefficients of Variation in Brackets

Composition	Flexural Modulus (E_{FL})	Tensile Modulus	Tensile Strength	As % E_{FL}
HMS	159 (7)	151 (15)	0.609 (11)	0.38
0.75 HTS 0.25 EHTS	138 (4)	129 (11)	0.604 (10)	0.44
0.5 HMS 0.5 EHTS	136 (7)	-	0.907 (10)	0.67
0.25 HMS 0.75 EHTS	118 (4)	105 (6)	0.936 (11)	0.79
0.25 HMS 0.75 HTS	113 (5)	108 (12)	0.996 (9)	0.88
EHTS*	99 (3)	97 (11)	1.156 (9)	1.16
HTS	100 (3)	114 -	1.14 -	1.14

*Revised (lower) surface treatment of HTS

TABLE 4

Interlaminar Shear Strength

of HTS-HMS Hybrids (50% Fibre)

Composition	ILSS (MPa)
HMS	67.8* (7)
0.75 HMS 0.25 EHTS	68.9 (7)
0.5 HMS 0.5 EHTS	81.4 (7)
0.25 HMS 0.75 EHTS	79.4 (13) 79
0.25 HMS 0.75 HTS	95.3 (5)
EHTS	88.7 (14)

*Some flexural failures

TABLE 5

Residual Tensile Strength of Crossplied Strips 1 mm Thick after Flexural Impact (Single Tests)

Composition	Unidirectional Flexural Strength GPa	Tensile Strength in GPa after Impact at						
		0.125 J	0.18 J	0.21 J	0.25 J	0.5 J	0.75 J	1.0 J
HMS	0.88 (6)	0.30 (99%)	0.27 (89%)	always broke				
0.5 HMS 0.5 EHTS	1.14 (8)				0.44 0.38 (91%)	0 0.42	0.003 0.006	- -
0.25 HMS 0.75 EHTS	1.27 (6)					0.46 0.47 (99%)	0.044 0.090	0.06 0.097
0.25 HMS 0.75 HTS	1.42 (6)					0.51 0.530 (105%)	0.12 0.11	0.07 0.11
EHTS	1.35 (4)					0.42 0.42 (72%)	0.11 0.11	0.13 0.10
HTS	1.25 (5)							

CV of flexural strength given in brackets, also percentage retention of crossply strength, estimated from unidirectional values.

TABLE 6

HMS-Glass Mixed and Layered Hybrids, Tested in Flexure (MBB)

Composition	Flexural Stiffness GPa	Flexural Strength MPa	Minimum Flexural Strain %
HMS	134	389	0.29
0.67 HMS 0.33 Glass	108 (97.9)	468 (385)	0.43 (0.39)
0.5 HMS 0.5 Glass	81.3 (83.4)	532 (436)	0.65 (0.52)
0.33 HMS 0.67 Glass	65.7 (65.0)	556 (443)	0.85 (0.68)
Glass	34.4	578	1.68

Data for layered hybrids in brackets

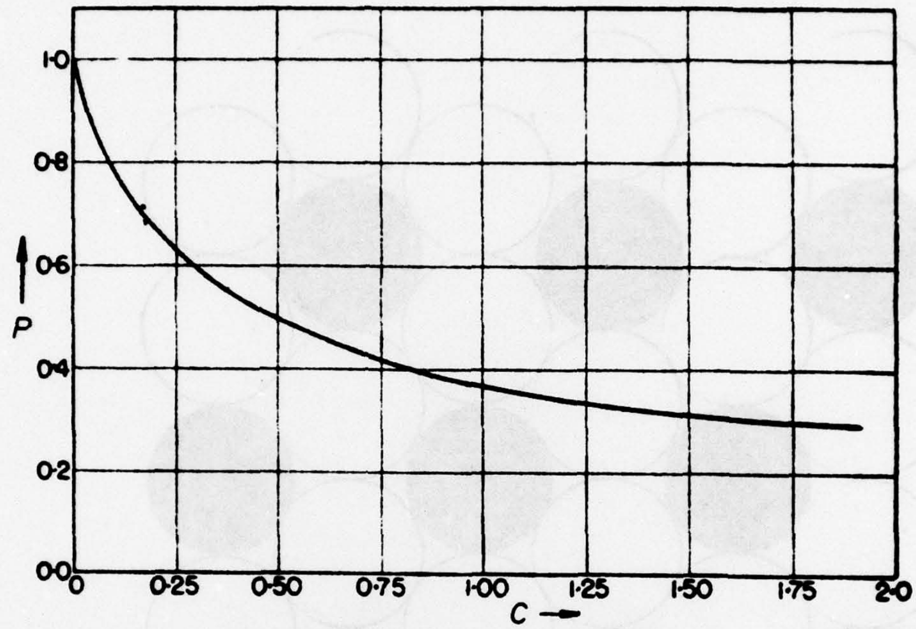

FIG. 1 THE STRENGTH EFFICIENCY P FOR AN INFINITE BUNDLE VS. THE COEFFICIENT OF VARIATION c IN THE STRENGTH OF THE COMPONENT FILAMENTS

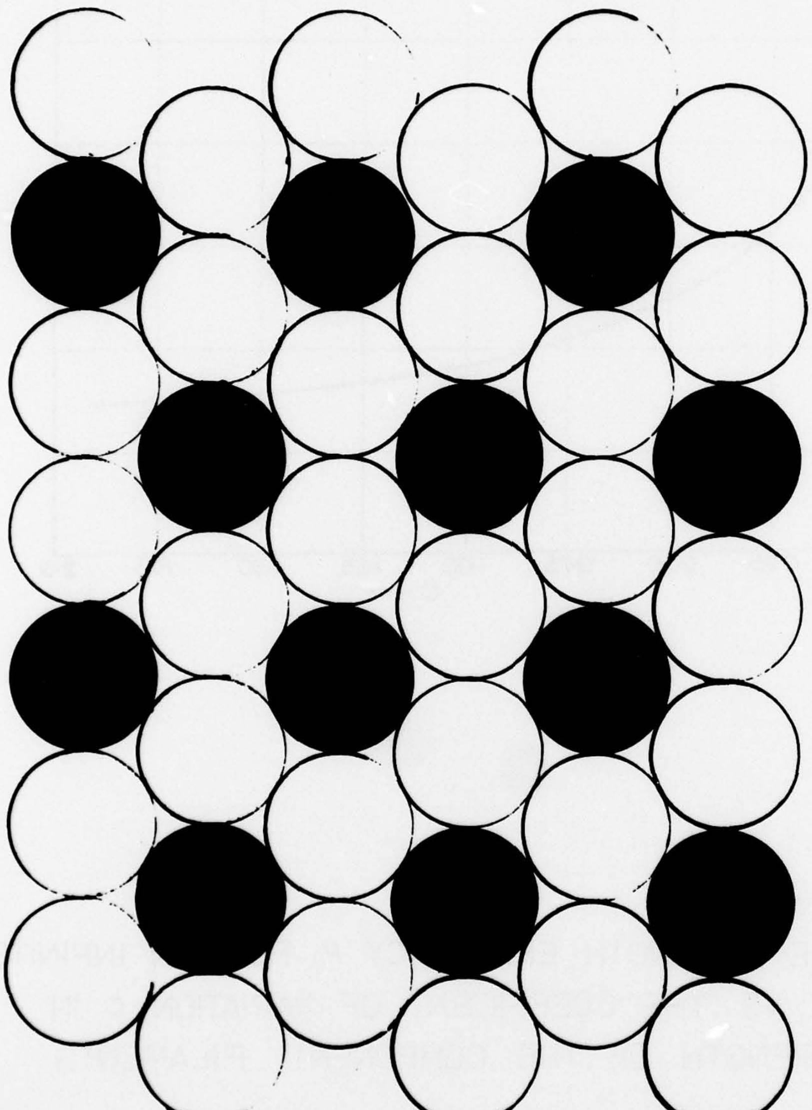

FIG. 2 ORDERED MIXING

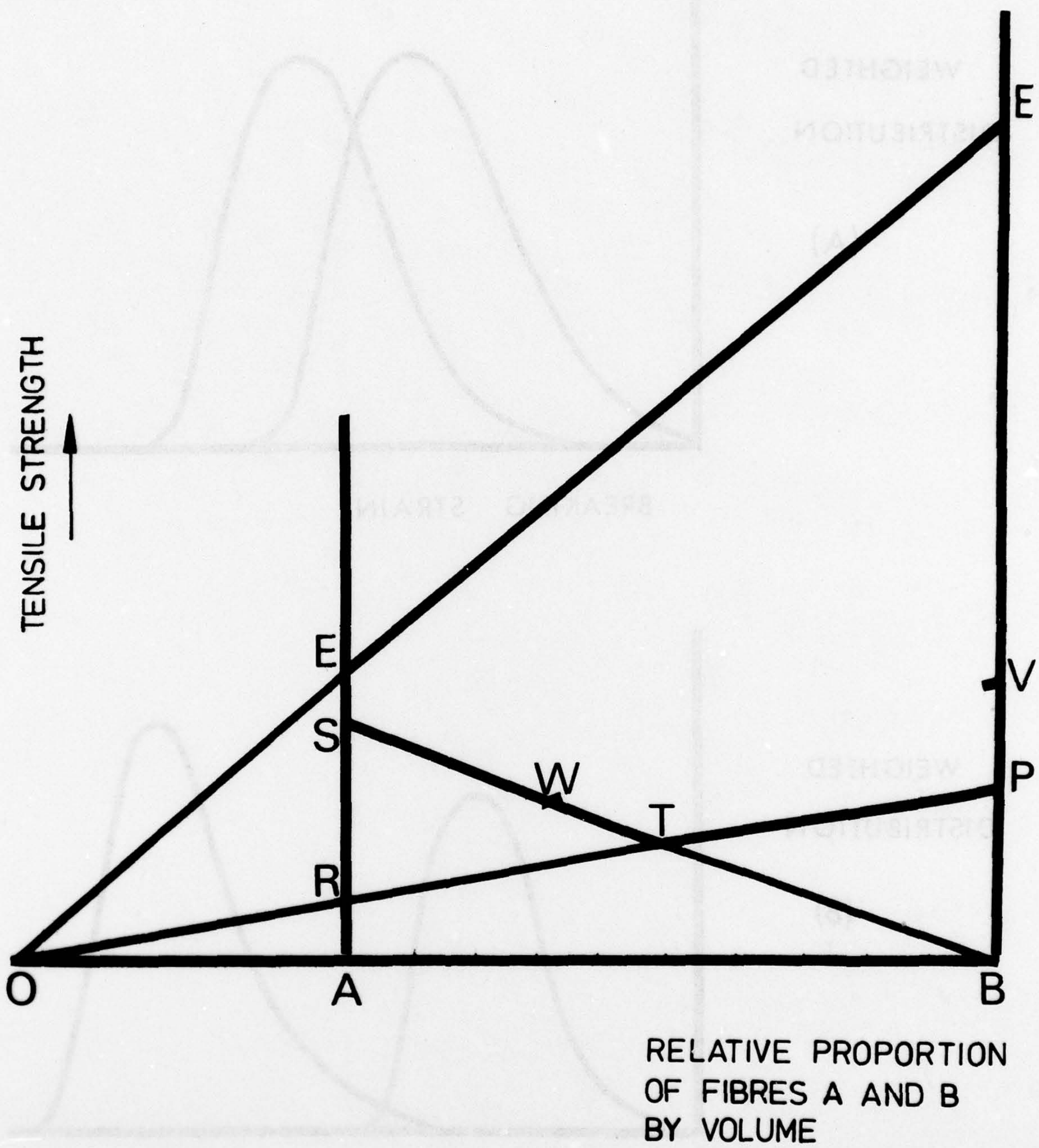

FIG. 3. STRENGTH AND STIFFNESS OF HYBRIDS CONTAINING FIBRES OR LAYERS (A AND B) OF LOW VARIABILITY

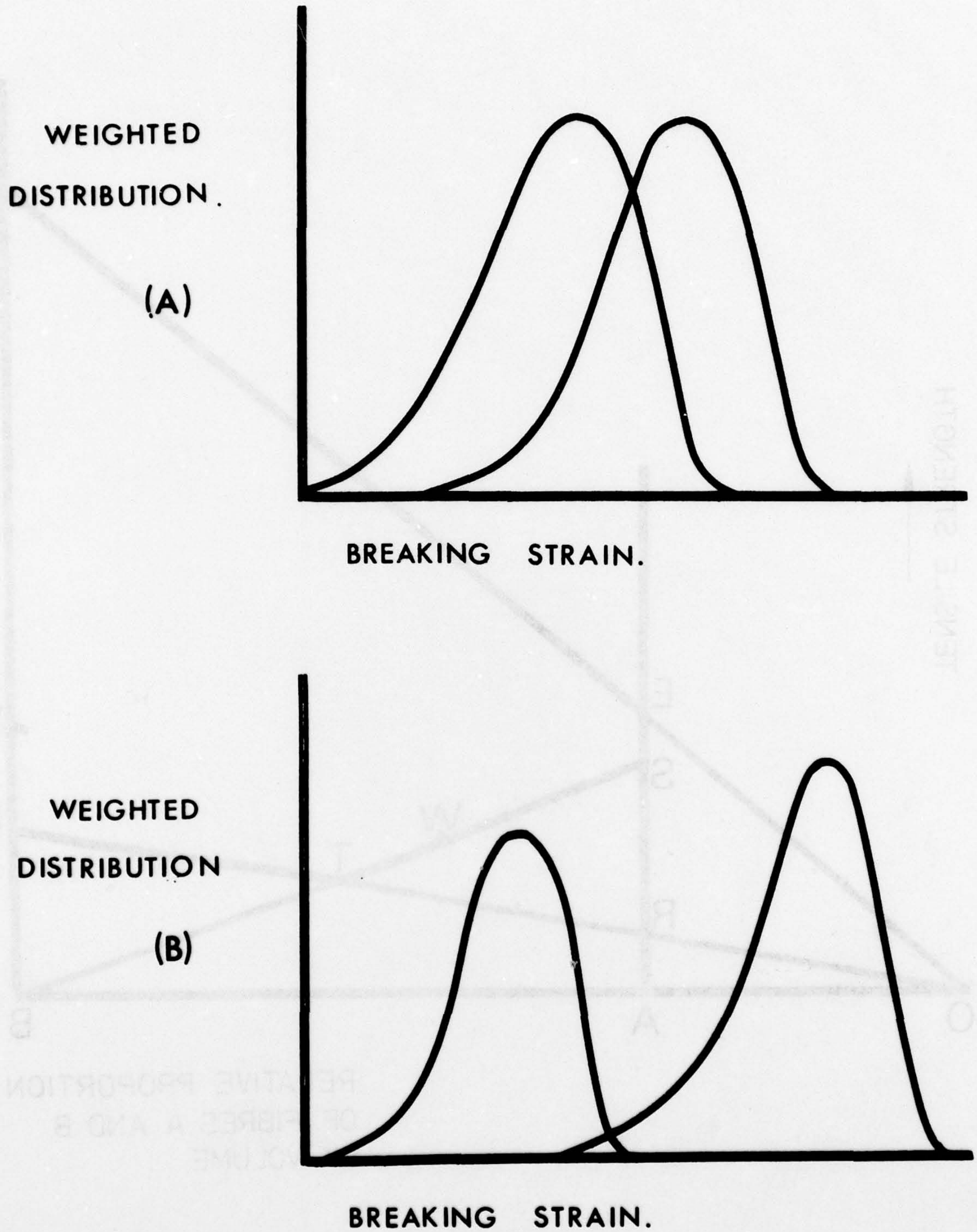

FIG.4 EFFECT OF OVERLAP WHEN COMBINING THE STRENGTH DISTRIBUTIONS OF TWO SETS OF FIBRES

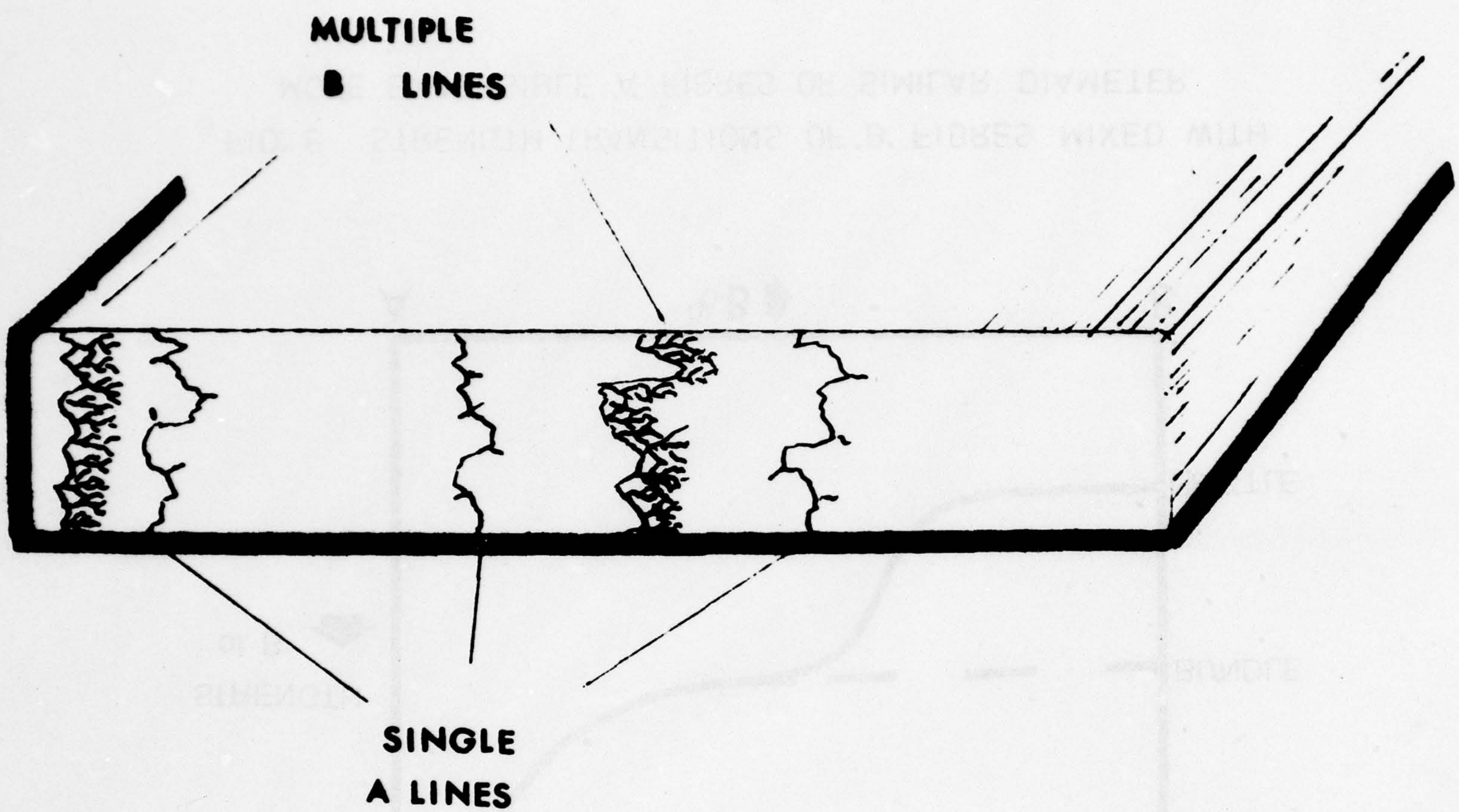

FIG. 5 SEQUENTIAL CRACKING OF 'B' FIBRES FROM A NOTCH OR INTERNAL PATH, WITH DECOUPLING LINES OF 'A'.

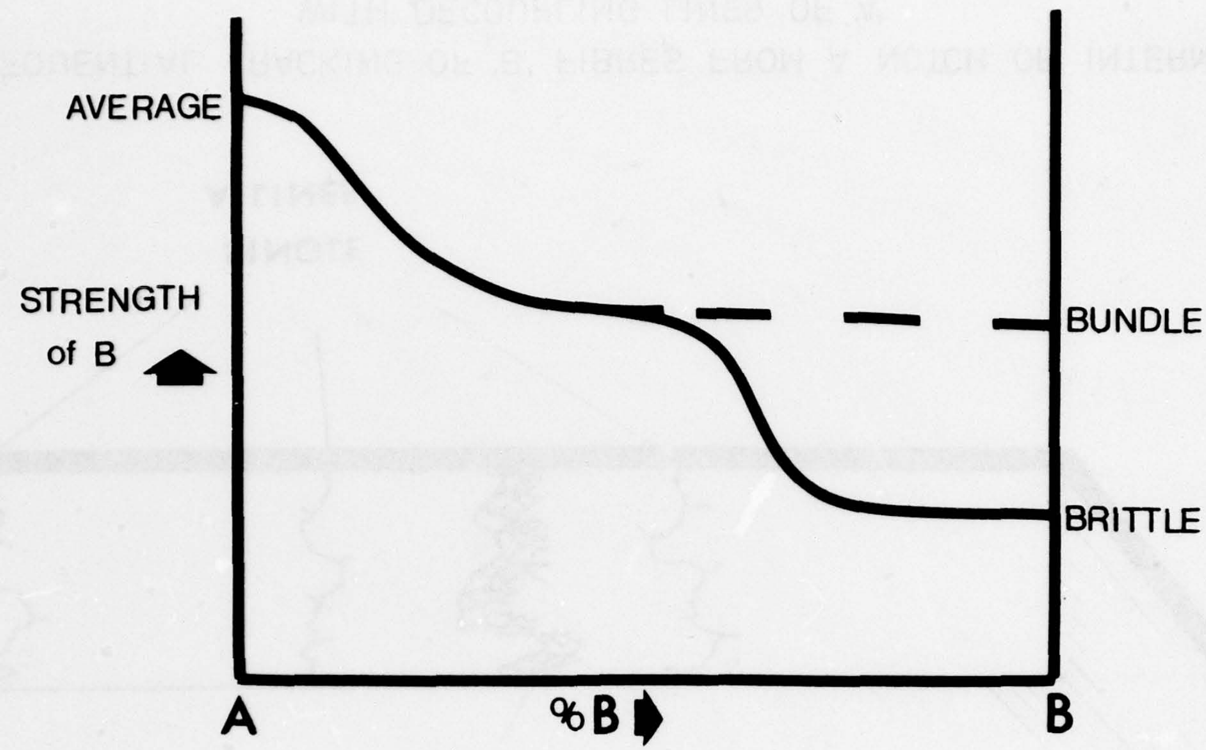

FIG. 6 STRENGTH TRANSITIONS OF 'B' FIBRES MIXED WITH MORE EXTENSIBLE 'A' FIBRES OF SIMILAR DIAMETER.

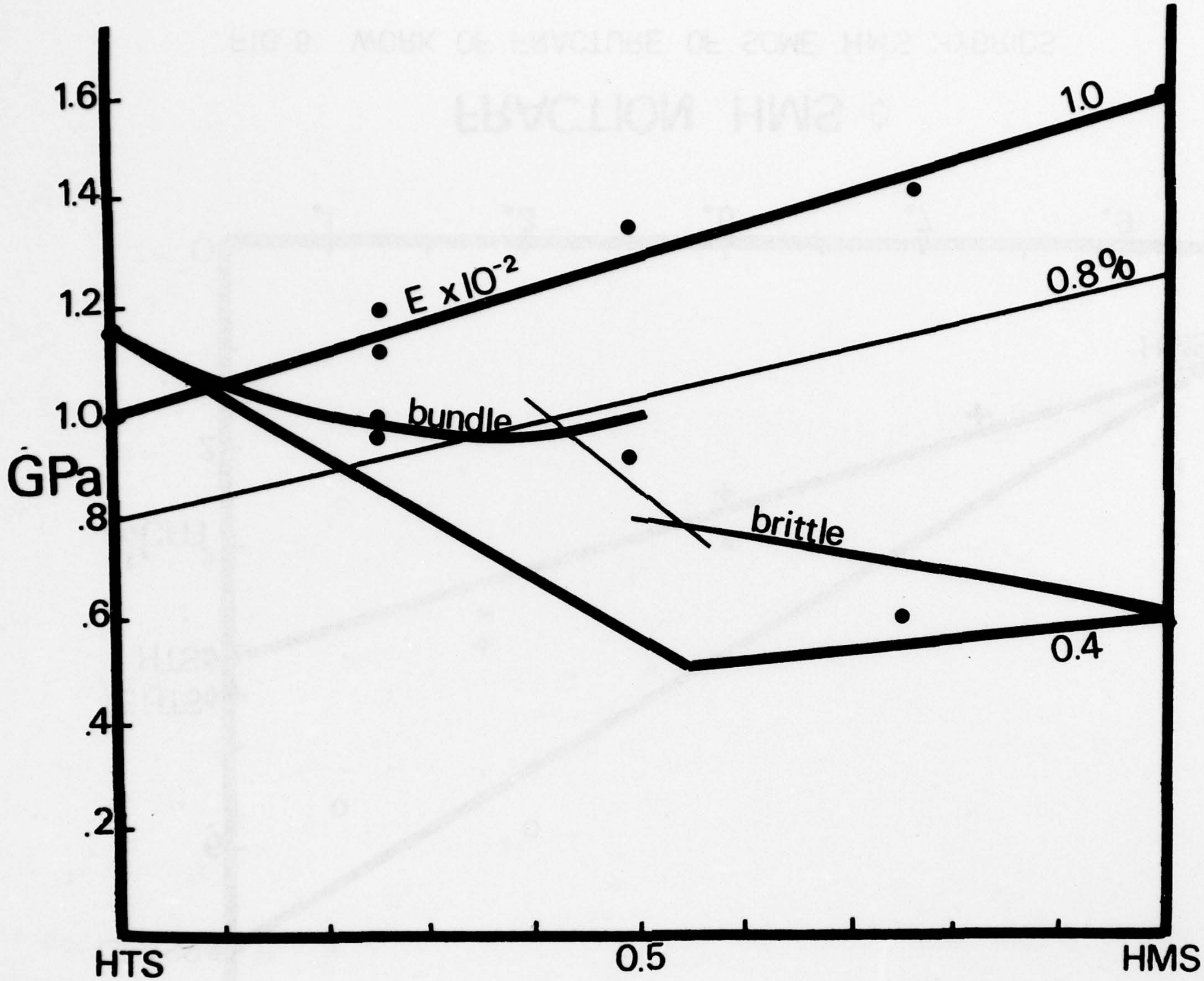

FIG. 7 OBSERVED TENSILE STRENGTH OF HTS-HMS HYBRIDS, WITH CONSTANT STRAIN AND STATISTICAL PREDICTIONS.

TR 81
FIG. 8

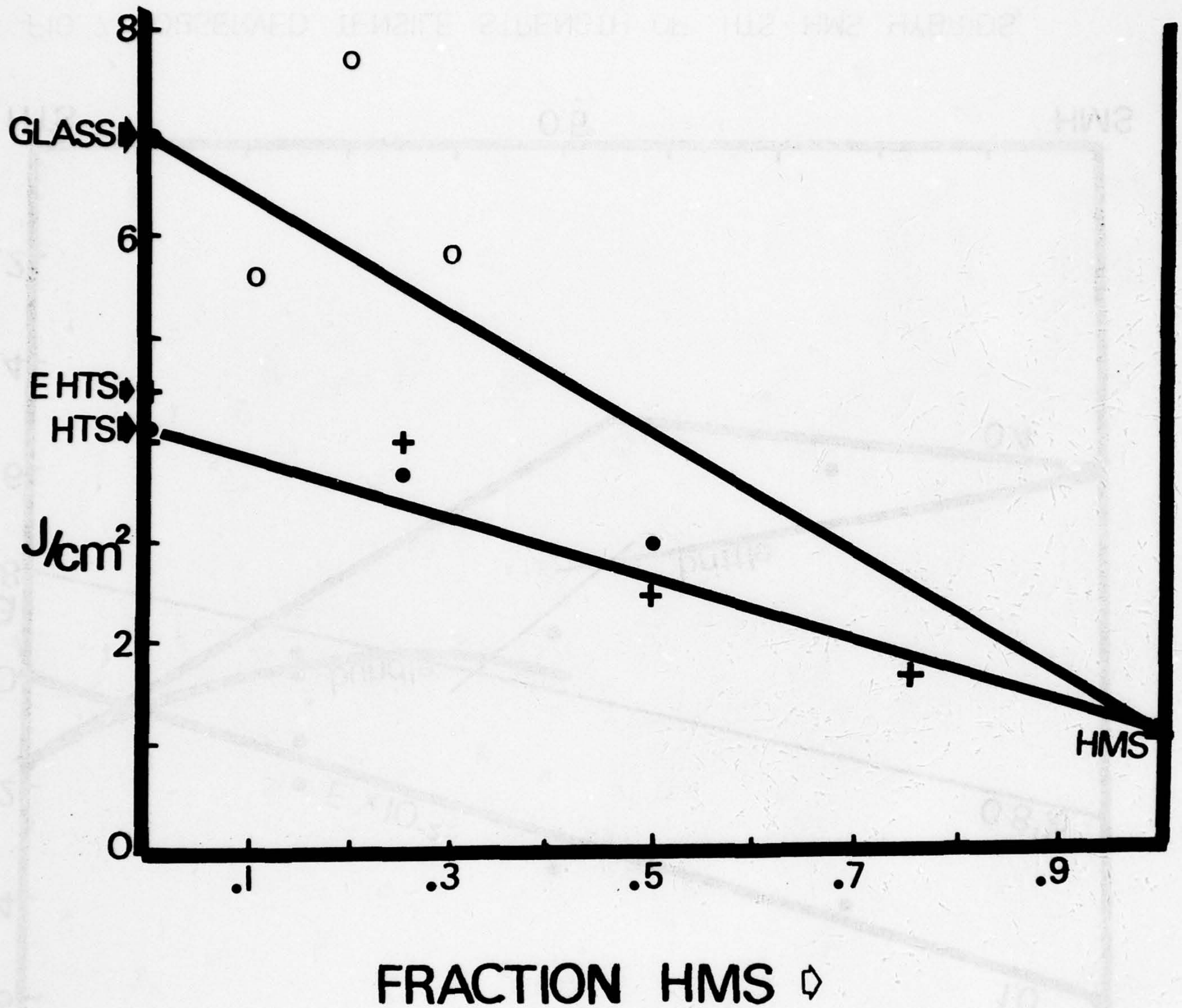

FIG. 8 WORK OF FRACTURE OF SOME HMS HYBRIDS

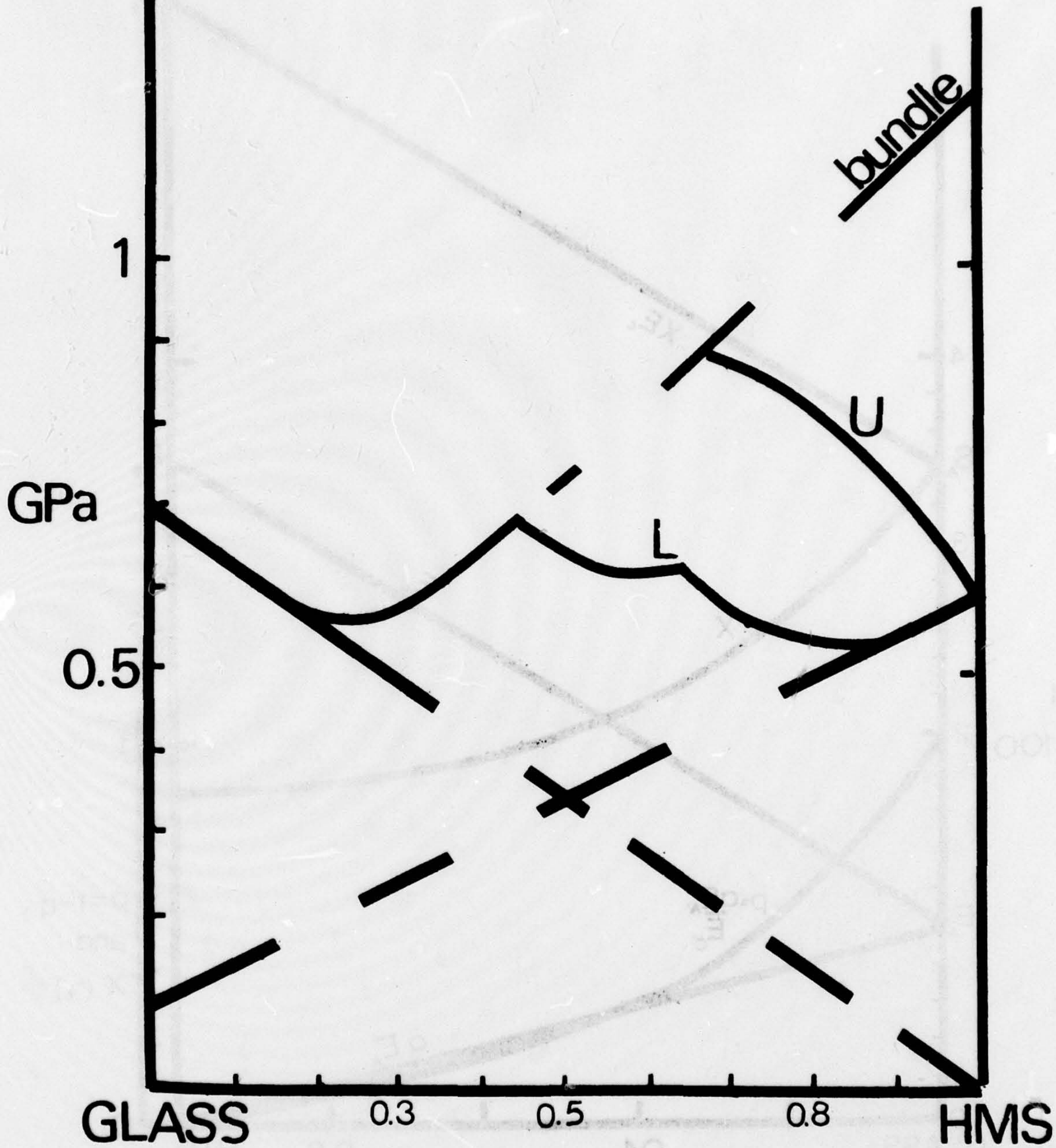

FIG. 9 SPECULATIVE ESTIMATE OF TENSILE STRENGTH IN THE GLASS - HMS SYSTEM

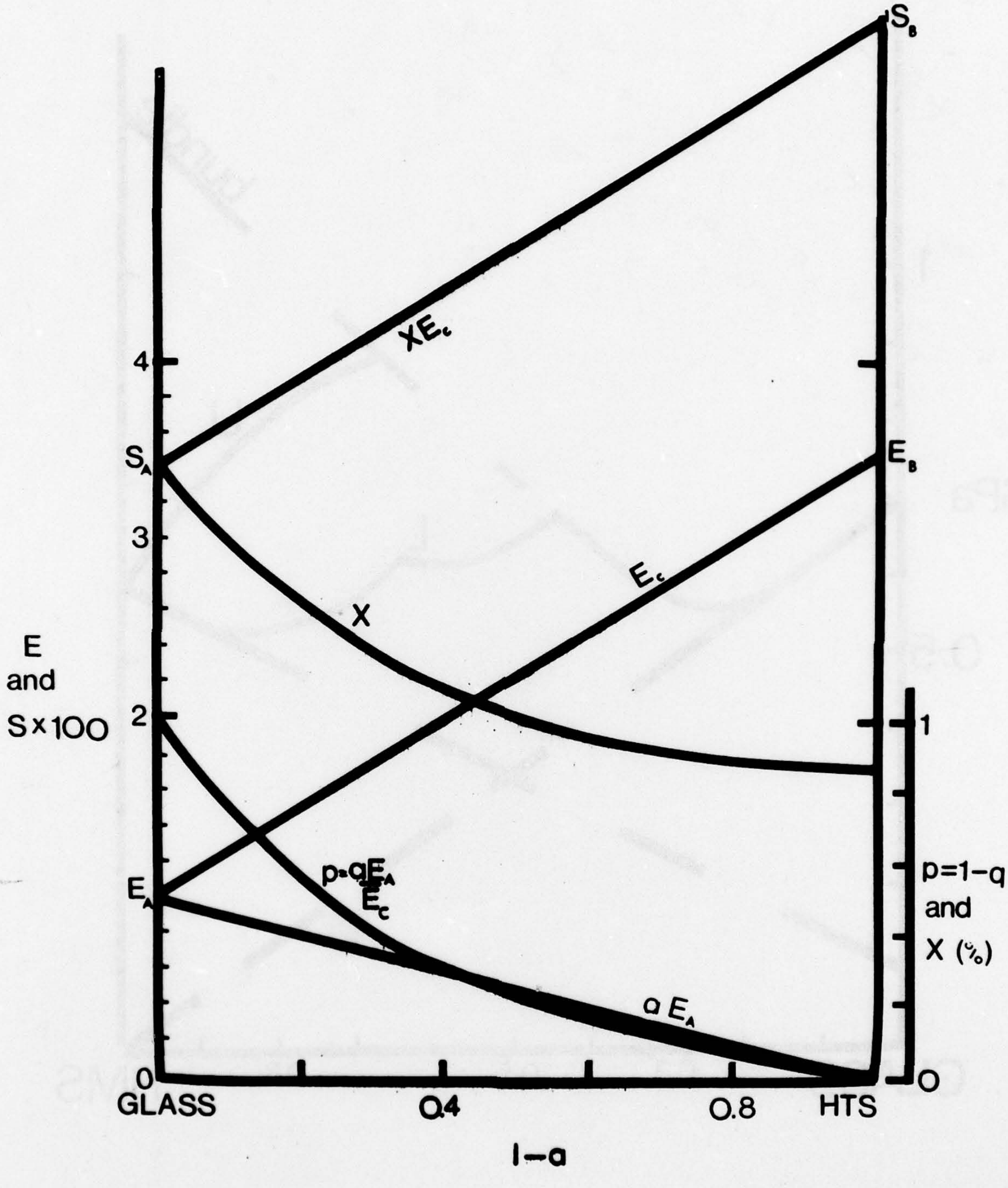

FIG. 10 GRAPHICAL ESTIMATES OF VARIABLES FOR CALCULATING HYBRID BUNDLE STRENGTH (SEE APPENDIX A)

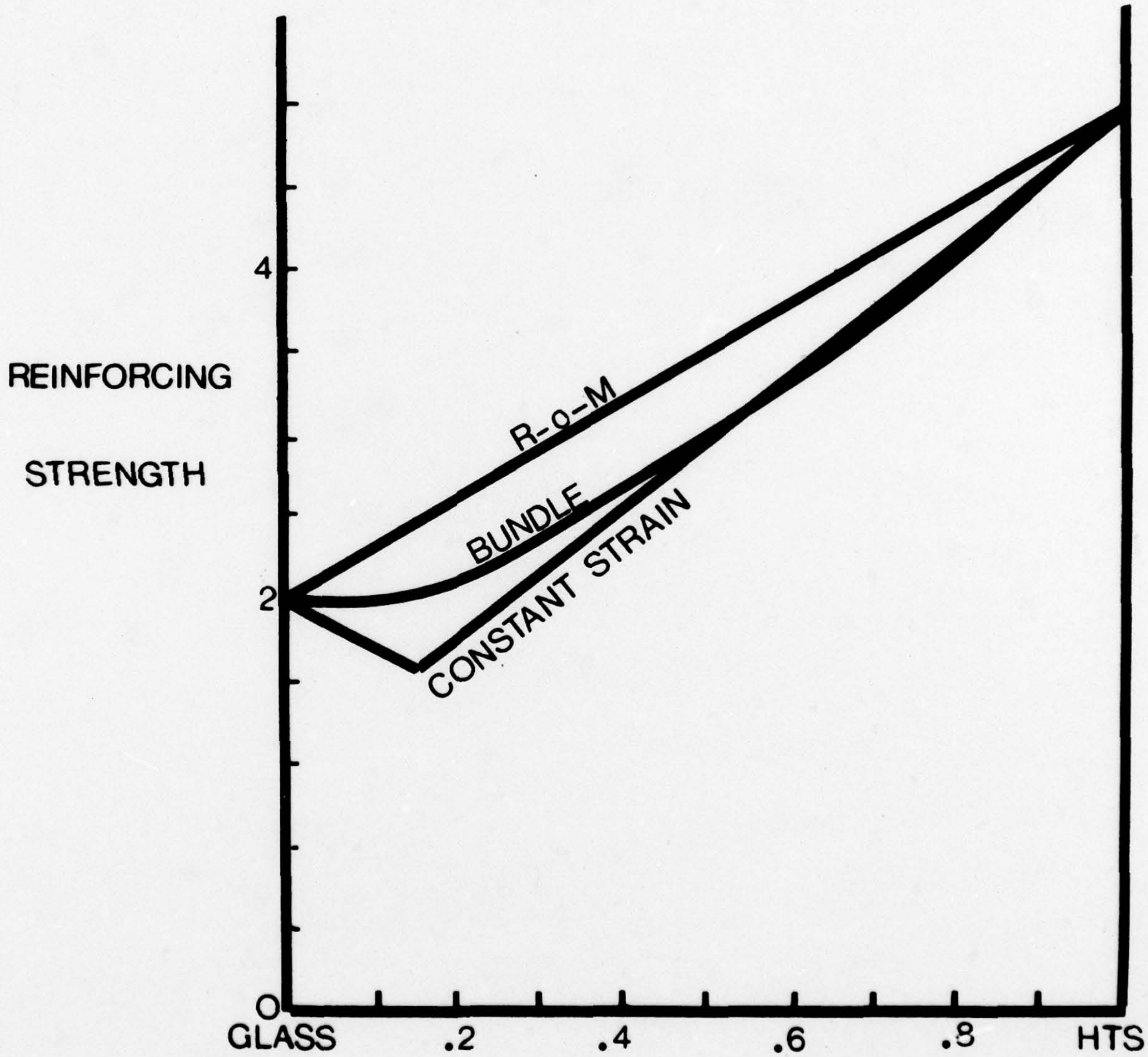

FIG. 11 CALCULATED REINFORCING STRENGTH OF GLASS - HTS FIBRES [UNITS 10^5 PSI (690 MPa)]

(Notes on completion overleaf)

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7b. Presented at (for conference papers). Title, place and date of conference

8. Author 1. Surname, initials	9a Author 2	9b Authors 3, 4...	10. Date	pp	ref
Parratt N J	Potter K D		11 78	40	15

11. Contract Number	12. Period	13. Project	14. Other References

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Descriptors (or keywords)

Chopped fiber composites, Alignment, Forecasting, Mechanical properties, Tensile strength, Carbon fibers, Glass fibers, Damage control

(TEST)

Abstract Starting from the constant strain theory of hybrid tensile strength, which only describes a lower bound for most hybrid composites, this report develops arguments to show how, in finely-mixed unidirectional hybrids the reinforcing strength of a set of fibres will assume higher values depending on their surroundings. In the case of hybrids of high modulus (HMS) carbon fibre, three characteristic levels of strength are predicted and indeed observed in the experiments reported here. These levels are, the mean fibre strength, the bundle strength referred to short gauge length, and the brittle strength which is also observed in all-HMS composites. Statistical co-ordination solutions are developed which predict the compositions of the average-bundle and the bundle-brittle transitions and also the hybrid tensile strength.

Recommendations are made for several practical systems. Those that have so far been investigated show the predicted trends, of which the most interesting are first,

DRIC/76/27

the use of HMS fibres with high tensile (HTS or Toray) carbon fibres to increase the stiffness and damage threshold of complex structures without serious loss of strength and second, the introduction of a glass fibre diluent which increases both work of fracture and strain to break, whilst lowering cost.